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13. ABSTRACT (Maximum 200 words)

This program supported the acquisition of instrumentation for the development of a system for coherent nonlinear optical spectroscopy of semiconductor heterostructures at liquid helium temperatures with sub-wavelength resolution. The design of the system is based on the super-resolution imaging technology developed for near field optical scanning microscopy (NSOM). The work included design and construction of an NSOM head capable of working reliably at low temperature with minimal vibration. Additional instrumentation acquired for this program was a low temperature cryostat with insert and associated electronics for NSOM control and signal acquisition. Special challenges in this work included the design and development of an all electrical feedback system to locate the tip relative to surface with improved performance over standard more cumbersome optical techniques. The system remains under development (supported by a separate grant from ARO), but at the time of this report, initial low temperature performance has been demonstrated.

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FINAL REPORT

Coherent Nonlinear Laser Spectroscopy Near Field Scanning Microscope

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A great deal of progress has been made in the effort to develop and build a low temperature near field optical microscope capable of performing nonlinear spectroscopy measurements. A dedicated laboratory has been acquired and outfitted. Low temperature, vacuum, and vibration isolation apparatus have been designed and built. An optical system capable of basic nonlinear measurements has been assembled and tested in the far field. A probe fabrication facility has been acquired and upgraded. And most importantly, a scanning probe microscope capable of functioning in high vacuum and at low temperature has been designed, built, run successfully under STP conditions, and is currently working in high vacuum.

The heart of the microscope lies with the long scan piezo. Due to the decreased responsivity at low temperature, the quadrant tube needed to be 2.5" long in order to achieve 10 μm travel in X and Y and 1 μm travel in Z. Titanium posts run the length of the microscope in order to compensate for the relatively low thermal contraction of the scan piezo and to ensure that the tip is not destroyed upon cooling down. The microscope is constructed largely of OFHC copper in order to keep thermal gradients to a minimum and thus to promote stability. The piezos are mounted on pieces constructed from MACOR, a machinable ceramic, because of its electrical insulating properties and its similar thermal contraction behavior. The sample stage allows for the mounting of a sapphire disk, with a transmission-optics plate below and a probe-plate above, both separately adjustable and independent of the sample stage. For mounting on the transmission plate, collection optics have been built with two aspheric lenses and a multi-mode fiber in a titanium housing in order to deliver light out of the cryostat. A photodiode or APD can also be mounted there. The coarse approach is operated remotely through a low temperature/vacuum compatible stepper-motor (Princeton Research), which, after being geared down, drives a lead screw into a differential spring arrangement attached to the sample stage. It can step over 500 μm in 30 nm steps. Coarse X and Y travel is also allowed for.

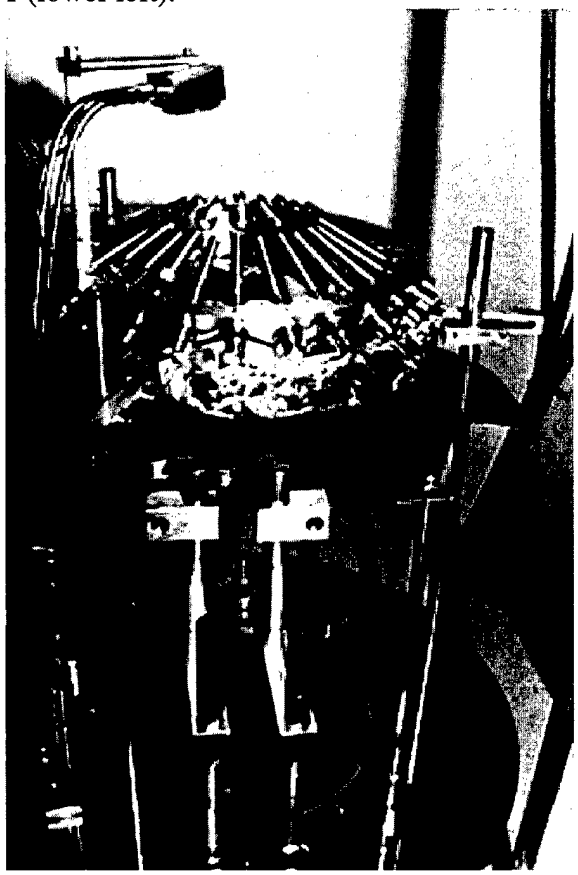
Due to the tremendously small distance scales associated with the scanning probe microscope, data integrity and probe health rely heavily on maintaining a stable tip-sample separation. In particular, the warming and cooling of the cryostat forces a 1-2 day hiatus if a probe is damaged and needs to be replaced. A great deal of effort, therefore, has been expended to minimize vibrational coupling into the tip-sample space. The laboratory that was acquired to build up this experiment was chosen largely because of its low mechanical noise baseline; it is located in the sub-sub-basement of the Randall Laboratory, i.e. 30 ft. below ground resting on grade and hundreds of feet from the nearest major street.

But a low vibration baseline is not enough. The microscope itself was designed to be extremely 'tight', i.e. the resonance frequencies of the microscope were set as high as possible. This was particularly difficult for a low temperature scanning probe microscope because of the reduced responsivity of the scanning piezo elements. In order to scan over respectable ranges, the elements needed to be much longer than their room temperature counterparts, pulling the resonance frequencies down. It was necessary, therefore, to invest in a 2-stage vibration isolation system. The first stage is a custom designed vibration isolation 'floating' workstation from Newport in which we mount the dewar. This workstation has vertical and horizontal resonance frequencies between 1.5 and 2.0 Hz, effectively blocking frequencies approaching the resonance frequencies of the microscope. The optional second stage is simply an undamped spring mounting of the microscope in the vacuum can at low temperature. This stage compounds the effectiveness of the first stage at high frequencies, but amplifies vibrations slightly at low frequencies.

In order to study samples at liquid He temperatures, it was necessary to design and build a dewar and an accompanying insert. The dewar was designed and built by Precision Cryogenics to our specifications. The insert was designed by ourselves and Janis and subsequently built by Janis. It suspends the microscope from a 1K pot in a high vacuum environment, allowing us to adjust the temperature of the sample down to 1.4 Kelvin. Numerous room temperature feed throughs allow access for unshielded wire, shielded wire (feedback signal), and optical fibers. The fibers include not only the single-mode fiber that is the near field probe, but also multi-mode fiber that returns light captured by the collection optics. By bringing the light back out of the cryostat, we open up the possibility of using a spectrometer for frequency resolution or an APD for extremely low light levels. Figure 1 (upper left) shows a photo of the NSOM assembly, without the sample stage and tip assembly attached.

Because of the inadequacy of commercially available NSOM tips, it was necessary to establish an in-house tip fabrication facility. We were able to acquire a previously owned P-2000 Sutter puller (designed specifically for pulling optical fibers) and Denton DV-502A evaporator in October

of 1997. While the fiber puller was quickly generating reproducible tips, the evaporator required some modifications and additions in order to coat the tips efficiently and effectively. The most important of these involved designing and building a 'tip rotator' that could spin 24 tips at ~12 Hz during the evaporation process. These modifications have been completed. A picture of the tip mount for the coater is shown in the Figure 1 (upper right) and a corresponding high resolution image of one of our uncoated tips is shown in Fig 1 (lower left).



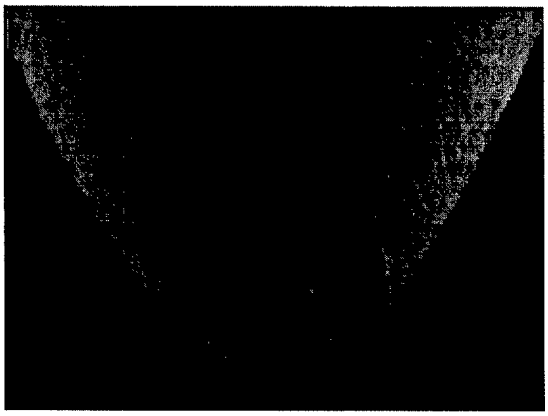


Figure 1. (Upper left) Photo of NSOM assembly. (Upper right) Tip holder assemble for the coater. (Lower left) SEM of tip. Total scale is 1200x950 nm.

Bringing the tip into the near field of the sample is particularly challenging in a low temperature/high vacuum environment. In addition to the lack of accessibility to the microscope

for adjustment (i.e. tip monitoring must be simple and robust), the vacuum environment denies the luxury of an interaction range and thus inhibits traditional analog feedback from maintaining a given tip-sample separation.

In addressing the first difficulty, it was deemed advisable to abandon the traditional optical feedback techniques and utilize a purely electronic approach. The microscope currently maintains its tip sample separation by feeding back on a 'piezo microphone' signal. While the tip is being driven at its resonance frequency by one quadrant of a 'dither' piezo, the opposite quadrant generates a signal that provides a strong and simple feedback signal. The primary modifications in the technique involve current rather than voltage detection, which helps to reduce the noise in our geometry, and phase sensitive detection. The latter improvement reduces the noise significantly, because the tip - when oscillating on resonance - generates a signal 90° out of phase with the driving signal. Therefore, by tuning the reference signal to 90° out of phase with the driving signal as well, the background can be reduced by at least another order of magnitude. This reduction is critical in order to obtain a accurate and fast feedback signal from the relatively low mass probe. In addition to the noise reduction, phase sensitive detection also provides a signal that approaches zero as the tip approaches the surface; this is extremely favorable behavior for feedback. A typical approach curve (at STP) can be seen in Fig. 2 below. Equally critical is the design of the dither piezo and probe section; it was kept as small and rigid as possible in order to keep the frequency spectrum clean below 30 kHz (we currently dither at ~ 20 kHz). This technique has been used extensively in our setup at room temperature, and has also seen success in vacuum.

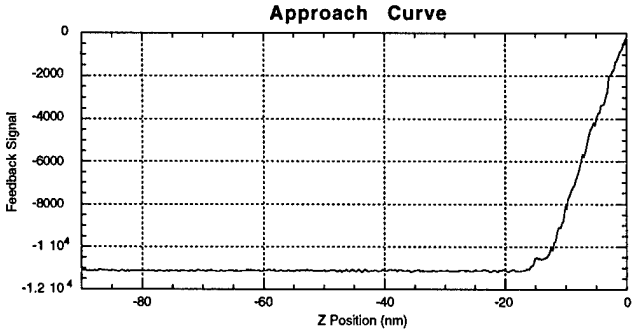


Figure 2. An approach curve at STP. Note the indication of a water bridge formation at the base of the approach.

In Fig. 2, it should also be noted that at the base of the approach, there is some indication of water bridge formation. In rough vacuum at room temperature the water bridge region has proven to be much shorter (<5 nm), and at low temperature even that source of damping will disappear.

There are multiple ways around this problem. The first is to take advantage of the relatively flat semiconductor samples and the extreme drift stability that low temperature affords and simply turn the feedback off. The second is to create our own 'interaction range' by tapping the tip on the surface and then rely on traditional analog feedback. The third is to implement a 'digital' feedback, which would involve reestablishing the location of the surface at each point on the surface. Our expectation is that the first solution will prove sufficient, but some work has gone into developing the alternatives.

Another issue that arises in vacuum is the change in the probe resonance; it shifts to higher frequency and sharpens, as can be seen in Fig. 3. While the shift is usually not a problem, the higher Q means a longer time response for the probe forcing slower scan speeds. In general, this should not be an issue for us as the photon collection rate will be the limiting step. It should also be noted in Fig. 3 that we have achieved a 17° and 38° phase shift in atmosphere and vacuum, respectively.

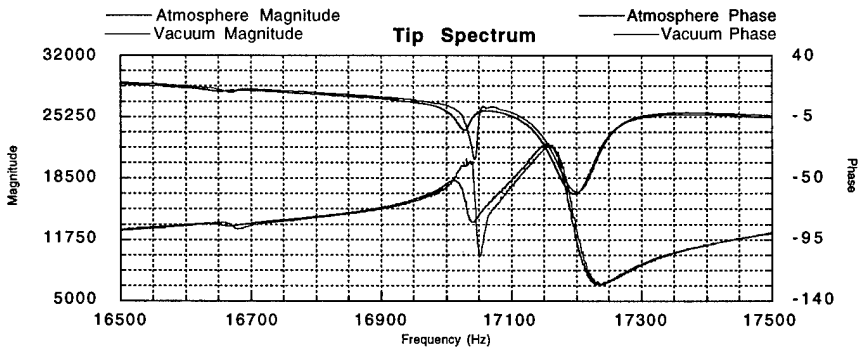


Figure 3. Resonance spectrum of the tip in atmosphere and at high vacuum. Note the shift to higher frequency and the sharpening of the tip resonance at 17045 Hz. Also note the absence of change to the other tip independent resonance.

While there are commercial software and electronics packages available to control scanning probe microscope systems, it was decided that for the cost they were too inflexible and limited for an experiment as ambitious as ours and incompatible with the rest of the laboratory. In was not clear, for example, that these commercial systems could manage to scan lasers, take data from CCD's, or move translation stages, without a great deal of extra 'macro' programming, if at all. In addition, the unique nature of the low temperature environment introduces some obstacles that may not be adequately addressed by a system designed for the majority of scanning probe microscopes operating in STP, most notably the feedback issue. Our entire laboratory is interfaced through LABVIEW on the Macintosh, so software has been developed on this platform to control the microscope that relies heavily on its flexibility and compatibility.

It should be noted that while the microscope has been designed, built, and tested in the dedicated laboratory alongside a newly outfitted optical table, it was constructed so as to be as mobile as possible. When the microscope is functioning reliably at low temperature, the possibility of moving it into the main laboratory to utilize the CW and time domain spectroscopic probes will be available to us.

While the instrument is not designed to perform extensive imagery, a basic test of performance is shown in Fig. 4 where we obtained a topographic image of a high contrast structure demonstrating the fundamental performance at low temperature in vacuum.

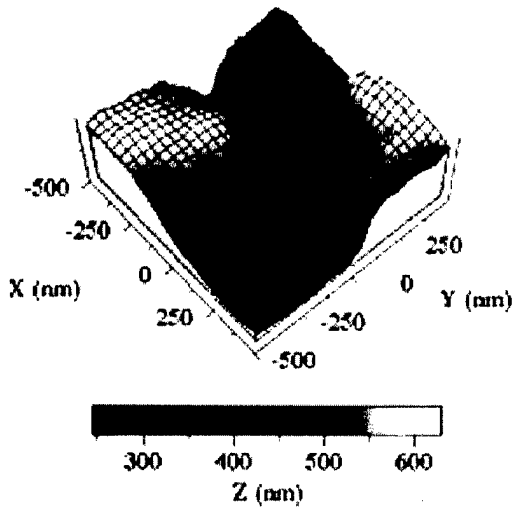


Figure 4. Topographic Image in vacuum at low temperature of a structured target showing fundamental performance.

Finally, in line with our work to develop improved performance in these systems for application to spectroscopy, we also demonstrated the feasibility of using two-photon excitation. The development was done using an NSOM developed separately for work at room temperature. The two-photon excitation approach was considered because of the need, in some cases, to work on extended objects. In these systems, optical signatures can be created away from the surface which then interfere with the surface optical emission and lead to a loss of resolution because of the rapidly diffracting optical field emerging from the fiber-optic probe. Using two-photon excitation rather than single photon excitation greatly reduces this problem because of the quadratic dependence of the excitation rate on intensity. We demonstrated the feasibility of this approach by using a 100 fsec Ti-Sapphire laser system to generate super resolution optical images of single dye molecules. Most impressive in this work is that we achieved super-resolution without metal coating of the tips. Earlier reports of super resolution without metal coating have been controversial since it appears that these data may be the result of topographical features that accompany the optical signatures. However, in the present work, we show that our surface is topographically free and the super-resolution is real. In fact, the resolution is in agreement with

simple theoretical description; the observation of 180 nm resolution for 800 nm excitation is approaching our estimate of the theoretical limit. The resulting single molecule image is shown in Fig. 5.

Figure 5. Image of single molecules obtained at STP based on two-photon induced fluorescence.